# DEPARTMENT OF THE INTERIOR U.S. FISH AND WILDLIFE SERVICE REGION 7

# ENVIRONMENTAL CONTAMINANTS PROGRAM ON-REFUGE INVESTIGATIONS SUB-ACTIVITY

Final Report

# AK-Mercury and Methyl-Mercury in Northern Pike (*Esox lucius*) from National Wildlife Refuges in Alaska

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by

Angela Matz Environmental Contaminants Program Leader

for

Sarah Conn, Field Office Supervisor Fairbanks, AK

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#### **ABSTRACT**

All National Wildlife Refuges in Alaska except Kenai include "continued use for subsistence activities" as a defined refuge purpose under the Alaska National Interest Lands Conservation Act. Contaminant burdens in fish and wildlife species used for subsistence can limit or preclude the continued opportunity for traditional subsistence activities on these refuges. Northern pike (Esox lucius) are heavily used subsistence foods on NWRs in Alaska, and have mercury concentrations that often exceed critical values for human consumption. However, few NWR managers in Alaska have data sets on mercury in pike and other fish that would allow them to manage for continued subsistence uses and to mitigate hazards from consumption of those fish. To address this lack of data, we conducted a multi-year, systematic sampling for mercury and methyl-mercury in northern pike on the largest refuges in western and interior Alaska: Yukon Delta, Selawik, Koyukuk, Nowitna, Innoko and Yukon Flats NWRs. From 2005- 2007, we collected 440 northern pike from these Refuges, in six major watersheds. Sample sites were traditional and well-used subsistence fishing locations, identified by local residents. Pike were collected during the time of year that they were normally collected for subsistence purposes, either in the spring (caught through the ice during spawning migration) or summer (caught in open water, post-spawning). Fish were necropsied and an approximately 10 g muscle sample was analyzed for mercury. Methyl mercury was analyzed in about a third of all samples. Mercury and methyl mercury were correlated in fish muscle, allowing calculation of a robust MeHg:THg ratio (0.96). Mercury was also correlated with fish length. We compared average calculated (based on the MeHg:THg ratio) methyl mercury concentrations from Refuges and watersheds to State consumption advisory values for mercury and found that some consumption advice was warranted, especially on the lower Yukon River in the Yukon Delta NWR. Mercury concentrations in pike muscle were of no apparent concern for fish health. These data will help Refuge managers provide continued subsistence opportunities and mitigate potential hazards associated with mercury in norther pike, a commonly used subsistence food. These data can also be used as baseline biomonitoring data for NWRs in Alaska, as mercury deposition, mobilization, and transport is likely to increase in light of increasing industrialization upwind of Alaska and in response to climate change.

#### **KEYWORDS**

Northern pike, Esox lucius, mercury, subsistence, Alaska, Yukon Delta NWR, Yukon Flats NWR, Selawik NWR, Innoko NWR, Koyokuk-Nowitna NWR Complex, Congressional District - Alaska #1, FFS: 7N24, DEC ID: 200570001.

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#### LIST OF ACRONYMS/ABBREVIATIONS

ANOVA – Analysis of Variance ANCOVA – Analysis of Covariance FDA – Food and Drug Administration ft - Foot FWS – Fish and Wildlife Service Hg - Mercury kg - Kilogram MeHg – Methyl mercury mg - Milligram NWR – National Wildlife Refuge PL – Public Law

ppm – Parts per million

SD – Standard Deviation

SE – Standard Error

THg – Total mercury

USEPA – U.S. Environmental Protection Agency

ww – Wet weight

#### INTRODUCTION

Most National Wildlife Refuges (NWRs) in Alaska were created, consolidated, or expanded under the Alaska National Interest Lands Conservation Act (PL 96-487, Dec. 2, 1980) which established the Department of the Interior responsibility for public safety relative to subsistence resources by stating, "...the Secretary...may temporarily close any public lands (including those within any conservation system unit), or any portion thereof, to subsistence uses of a particular fish and wildlife population only if necessary for reasons of public safety, administration, or to assure the continued viability of such population." All NWRs in Alaska except Kenai include "continued use for subsistence activities" as a defined refuge purpose. Contaminant burdens in fish and wildlife species used for subsistence can limit or preclude the continued opportunity for traditional subsistence activities on these refuges.

Northern pike (*Esox lucius*) from Alaska contain mercury concentrations that often exceed the U.S. Environmental Protection Agency (USEPA) critical value for human consumption of 0.3 milligrams/kilogram (mg/kg) wet weight (ww) (USEPA 2001a, b) and the U. S. Food and Drug Administration (FDA) action level for human consumption of fish tissue (the concentration at which fish sales are restricted) of 1.0 mg/kg ww. Mean mercury concentrations in northern pike from Alaska and Arctic Canada equal or exceed those from the Lower 48 United States (USEPA 1992). For example, eight of nine northern pike from Nowitna NWR in 1987 and six of nine in 1991 had concentrations in tissues exceeding the EPA critical value (Snyder-Conn et al. 1992, Mueller et al. 1996). More recently, eight of nine northern pike examined from the Andreafsky River, a tributary on the lower Yukon River, had mercury concentrations in muscle that exceeded the FDA action level (Duffy et al. 1999; Jewett et al. 2003).

Although mercury data in pike from NWRs in Alaska were collected from 1987 - 2000, these collections represent small numbers and limited sampling areas. Refuges in western and interior Alaska (Kanuti, Innoko, Koyukuk, Nowitna, Selawik, Tetlin, Yukon Flats, and Yukon Delta NWRs) total greater than 16 million hectares (41 million acres), but the existing database was numerically and spatially depauperate. To address this lack of data, which Service managers need to make subsistence management decisions, we conducted a multi-year, systematic sampling for mercury and methyl-mercury in northern pike on the largest refuges in western and interior Alaska: Yukon Delta, Selawik, Koyukuk, Nowitna, Innoko and Yukon Flats NWRs.

These Refuges were chosen for this project because they are large, have many subsistence users, and northern pike are commonly used as a subsistence food. In the spring fresh northern pike are a welcome and substantial addition to the winter diet of dried salmon for the Yupik people. In some villages, and for some families in all villages, northern pike represent a year-round protein source. Further, Jewett et al. (2003) stated that mercury concentrations in northern pike from some Yukon Delta rivers were high enough that consumption of large amounts of northern pike was hazardous for all age groups. Northern pike also are an important protein source for some families in villages within Yukon Flats and Selawik NWRs, and managers from both refuges have expressed specific interest in understanding mercury issues for subsistence management.

The few data available for these two refuges indicated lower mercury concentrations in pike compared to Yukon Delta NWR (U.S. FWS unpubl. data), but sample sizes were small and insufficient for making subsistence management decisions.

The primary goal of this study was to create a substantive data set on which subsistence management and consumption decisions could be based for northern, western and interior Alaska NWRs. Ancillary goals include testing for correlation between mercury and methyl mercury and establishing an average methyl mercury:total mercury (MeHg:THg) ratio in northern pike muscle; establishing the average northern pike length above which muscle mercury concentrations are likely to exceed consumption thresholds; determining differences among watersheds for mercury in pike; and determining if mercury concentrations are at concentrations that may interfere with northern pike reproduction or development by comparison with literature values.

#### **METHODS AND MATERIALS**

#### Field Methods

From 2005- 2007, we collected 440 northern pike from 32 sites on six National Wildlife Refuges (Fig. 1) and six major watersheds in northern and western Alaska (Table 1). Sample sites (Appendix A) were traditional and well-used subsistence fishing locations, identified by local residents. Pike were collected during the time of year that they were normally collected for subsistence purposes, either in the spring (caught through the ice during spawning migration) or summer (caught in open water, post-spawning). Pike of sizes usually eaten were collected with hook and line or with gillnets. After collection, pike were humanely dispatched and frozen until shipment to the FWS laboratory at the Fairbanks Fish and Wildlife Field Office in Fairbanks.



Figure 1. National Wildlife Refuges in Alaska where northern pike (*Esox lucius*) were collected for mercury and methyl-mercury analysis, 2005-2007.

Table 1. Number of northern pike (*Esox lucius*) collected for mercury and methyl-mercury analysis from National Wildlife Refuges in Alaska, 2005-2007.

National Wildlife Refuge	Watershed Region	Number of pike
Yukon Flats	Upper Yukon River	70
Yukon Delta	Kuskokwim	103
Innoko	Mid-Yukon	47
Yukon Delta	Lower Yukon	86
Koyukuk/Nowitna	Mid-Yukon	30
Selawik	Northwest Alaska	104

#### Laboratory Methods

At the lab, pike were necropsied and muscle samples for total and methyl mercury analysis were collected following established protocols (Schmitt et al. 1999, Smith et al. 2002). Briefly, fish were weighed, measured (fork and total length), internal and external health assessments were performed, otoliths were collected for aging, and an approximately 10 g sample of skinless filet (muscle) was collected anterior to the left gills and dorsal to the lateral line, using chemically clean instruments. Muscle samples from all fish were placed in chemically clean jars appropriate for metals analysis and were analyzed for total mercury (Hg); muscle samples from 146 fish (1/3 of total) were placed in chemically clean jars appropriate for organics analysis and were analyzed for methylmercury (MeHg). All analyses were performed at the Trace Element Research Laboratory at Texas A & M University, College Station, TX, using standard methods.

#### Statistical Methods

General linear models, similar to Analysis of Variance (ANOVA) and Analysis of Covariance (ANCOVA), were used to explore patterns and test differences in average mercury concentrations among and between sites, with a significance level set at  $\alpha = 0.05$ . We tested for correlation between fish length or weight and mercury concentrations; if significant, either length or weight was used as a covariate when either was significantly correlated with total mercury within a species. Post-hoc tests (Tukey's or Bonferroni's) were used to determine statistical significance for differences among sites.

#### **RESULTS**

Methylmercury and mercury were significantly correlated

Mercury and MeHg in northern pike muscle from Alaska were significantly and highly correlated (Pearson's correlation coefficient = 0.985, n=146). Most of the total mercury in northern pike muscle was the most toxic form, MeHg (mean  $\pm$  SD MeHg:THg = 0.963  $\pm$  0.050). We multiplied total mercury wet weight concentrations by the average MeHg:THg ratio to give a calculated MeHg wet weight concentration, which is how consumption limits are often depicted. We used this calculated MeHg value, usually log-transformed to reduce the influence of outliers, in all further analyses.

#### Fish size was correlated with MeHg concentrations

As expected, MeHg concentrations were significantly correlated with both fork length and weight (Pearson's r = 0.529 and 0.549, respectively; Bonferroni p < 0.001 for both). Fork length and fish weight were therefore used as covariates, when appropriate, in subsequent analyses. We also used the arbitrary but easily remembered length of 2 ft as the division between "small pike" and "large pike" for calculating consumption limits.

### MeHg concentrations

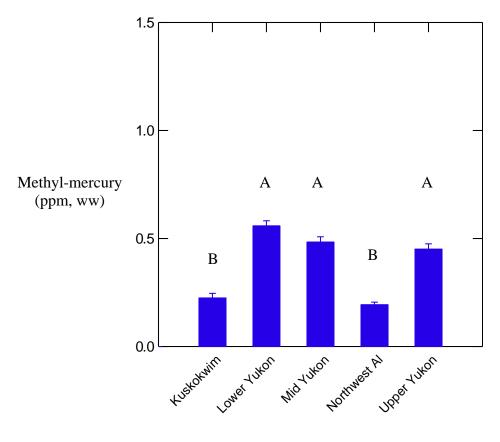
We calculated mean MeHg concentrations for all samples collected on each National Wildlife Refuge, to give each Refuge manager a Refuge-specific value (Table 2).

Table 2. Methylmercury concentrations (ppm, ww) in two size classes of northern pike (*Esox lucius*) skinless muscle from National Wildlife Refuges in Alaska, 2005-2007.

	National Wildlife Refuge				
	Yukon Yukon		Koyokuk-		
	Flats	Delta	Innoko	Nowitna	Selawik <sup>1</sup>
Pike shorter than 2 ft					
N of cases	39	98	8	13	81
Mean	0.346	0.278	0.364	0.370	0.172
Std. Error	0.027	0.021	0.067	0.067	0.012
Pike longer than 2 ft					
N of cases	29	65	37	17	23
Mean	0.587	0.605	0.580	0.387	0.268
Std. Error	0.031	0.026	0.031	0.034	0.025

<sup>&</sup>lt;sup>1</sup> Some samples associated with Selawik NWR were collected outside of Refuge boundaries.

Because mercury concentrations in fish can be related to watershed characteristics, including wetland areas, fire history, and local or point sources from underlying geology or mining activity, we used large watershed areas (Appendix A), rather than Refuge boundaries, as the units among which we tested for differences in MeHg concentrations. When not separated into size class, the Kuskokwim and Northwest Alaska pike had significantly lower MeHg concentrations compared to all three Yukon River watershed areas (ANCOVA, p < 0.001). However, there was a disproportionate number of small pike (n = 71) compared to large pike (n=6) from the Kuskokwim watershed. When viewed by size class, large pike from the Kuskokwim showed MeHg concentrations similar to large pike from the Yukon River, while small pike maintained the pattern seen among all pike combined (Fig. 3).



Watershed Region

Figure 2. Mean (+ SE) methyl-mercury concentrations (ppm, ww) in northern pike (*Esox lucius*) skinless muscle from northern and western Alaska, 2005-2007. Unlike superscripts indicate significant differences.

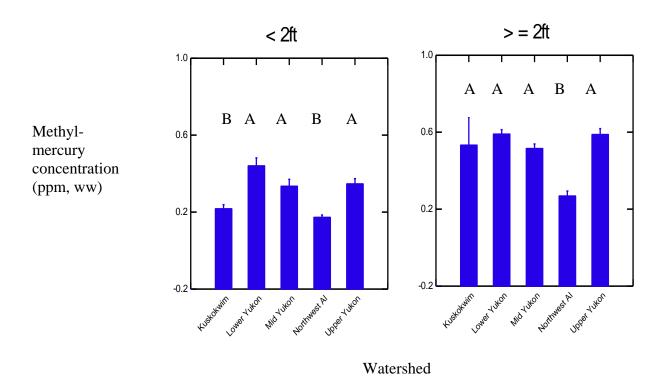


Figure 3. Mean (+ SE) methyl-mercury concentrations (ppm, ww) in northern pike  $(Esox\ lucius)$  skinless muscle from northern and western Alaska, 2005-2007. Data on the left are from pike less than two feet in length; data on the right are from larger pike. Large pike from the Kuskokwim have MeHg concentrations similar to large pike from the Yukon River areas; small Kuskokwim pike do not. Unlike superscripts indicate significant differences.

#### Mercury concentrations related to consumption guidelines

We worked with the State of Alaska Department of Health and Social Services, Division of Public Health, Epidemiology Section (State) to compare mercury concentrations in sampled pike to State consumption guidelines (Verbrugge 2007), which applied to women of childbearing age and children. We then prepared posters and other outreach materials that were specific to each region (e.g. Fig. 4, which shows the most restrictive guidance, from the Lower Yukon watershed region). We presented those materials at joint FWS-State public meetings in villages where we sampled pike and at regional hubs such as Kotzebue, Bethel, Galena, and Fairbanks.

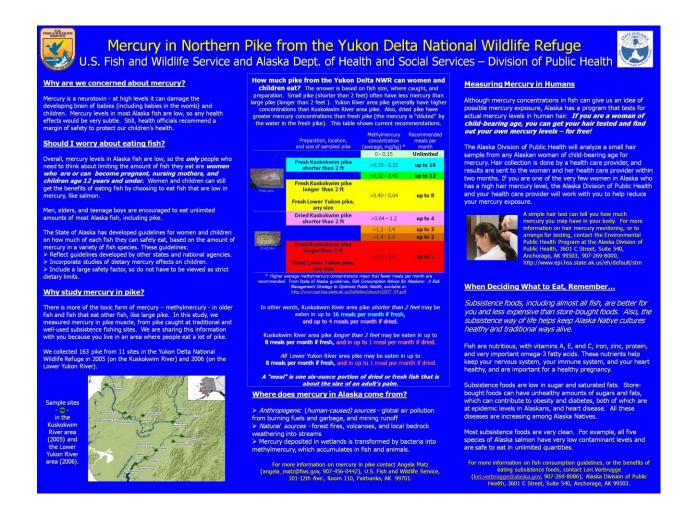


Figure 4. Example of outreach poster showing region-specific guidance for subsistence consumption of pike with methylmercury concentrations of concern from Alaska, 2005-2007.

There were guidance restrictions for pike consumption in all areas sampled, although the recommendations varied by region. Heavy emphasis was placed on choosing other healthy subsistence foods, such as salmon, which provide equivalent or better nutrition compared to pike, and the social and economic benefits of subsistence fishing.

#### DISCUSSION

The concern for mercury in northern pike in Alaska stemmed from their widespread use as a subsistence food. However, we also compared methylmercury concentrations in these pike to literature thresholds to determine if there were potential fish health impacts. The average methyl mercury concentrations documented in this study (Fig. 2, Fig. 3) were orders of magnitude lower than the 8.4 – 24 ug/g ww mercury in muscle for multiple fish species affected by mercury in Minimata Bay, Japan (Wiener et al. 2003). Pike with muscle mercury of 6-16 ug/g ww in a mercury-contaminated lake in Ontario showed clinical and overt signs of starvation, from which they recovered after being placed in a less contaminated lake (Lockhart et al. 1972, as discussed by Wiener et al. 2003). Other concentrations of concern for mercury or methyl mercury in axial muscle of fish range from 5-20 ug/g ww for overt toxicity (Wiener et al. 2003). Reproductive effects might occur at much lower concentrations, however, although waterborne mercury has had greater embryotoxicity to fish embryos compared to maternally derived methyl mercury in yolk (Latif et al. 2001). Currently, northern pike population declines in Alaska are not of concern, and this data set indicates that mercury in these fish is likely not a significant fish health threat.

Our data will also serve as baseline data for biomonitoring of mercury on National Wildlife Refuges in Alaska. While North American mercury emissions have declined with time, overall global mercury emissions are projected to rise as more coal-fired facilities are built in Asia, a region directly upwind of the Bering Sea, Alaska and the Western Arctic. In 2000, the largest emissions of mercury to the global atmosphere occurred from combustion of fossil fuels, mainly coal in utility, industrial, and residential boilers. As much as two-thirds of the total emission of approximately 2190 tons of mercury emitted from all anthropogenic sources worldwide in 2000 came from combustion of fossil fuels (Pacyna et al. 2006). Emissions of mercury from coal combustion are between one and two orders of magnitude higher than emissions from oil combustion, depending on the country. Asian countries contributed about 54% to the global mercury emissions from anthropogenic sources in 2000. China had the highest mercury emissions from anthropogenic activities, contributing about 28% of the global emissions of mercury in 2000 (Pacyna et al. 2006).

Climate change may also contribute to increased mercury mobilization, transport, and availability in Alaska. For example, sampling of peat cores from a Swedish mire and nearby lake sediment cores show that during past periods of warming (e.g., 1400's to mid-1500's), lake sediment mercury concentrations increased due to transport of organic matter and mercury (Rydberg et al. 2010). Current thawing of permafrost is again increasing themokarst erosion,

resulting in a wetter mire and transport of mercury to the lake. Rydberg et al. (2010) concluded that while these observations are from a single site, climate-related erosion of organically rich mires have the potential to influence mercury levels in other freshwater ecosystems within the subarctic and Arctic and mercury dynamics should be studied in other systems. Similar observations have been made for lead in subarctic Sweden, with increased transport of lead to lakes in areas experiencing permafrost melt, and lesser transport in boreal or birch-forest catchment lakes (Klaminder et al. 2010).

The fire data record for Alaska suggests an increase in large fires in response to recent climate warming (ACIA 2005). While the record is insufficient to determine definitively whether the increase is outside the range of natural variability, various models predict that a warming climate will lead to more and larger fires, and that warmer and wetter scenarios will produce more very large fires compared to the warmer and dryer scenarios. The warmer and dryer climate scenario resulted in more frequent medium-sized fires, which prevented fuels from building up across the landscape and limited the number of large fires (ACIA 2005). Recent data show a significantly increasing trend in total area burned within Alaska and Canada over the past 30 years, and both fire frequency and severity are expected to increase in northern boreal forests with continued climate change (Flannigan et al. 2009). Friedli et al. (2009) suggested that increasing temperatures in boreal regions, where the largest global soil mercury pool resides, will exacerbate regional and global mercury emissions due to more frequent, larger, and more intense wildfires.

The association of mercury emissions with wildfires is well studied. For example, Turetsky et al. (2006) found that mercury formerly sequestered in cold, wet peat soils was released to the environment during fires in Canadian boreal forests, which presented a growing threat to aquatic habitats and northern food chains as the climate warms. Estimates of circumboreal mercury emissions presented in this study are 15-fold greater than estimates that did not account for mercury stored in peat soils. Boreal areas with greater peatland abundance are correlated with the size of large fire events (Flannigan et al. 2009). Weidinmeyer and Friedli (2007) estimated mercury emissions from wildfires in Alaska and the lower 48 States from 2002-2006 to average 44 metric tons per year, roughly equivalent to 30% of the total mercury emissions permitted by the US Environmental Protection Agency in 2002. In 2004, a high fire year, fire-related mercury emissions in Alaska (32 metric tons) exceeded the combined total amount from the lower 48 states (23 metric tons) (Weidinmeyer and Friedli 2007). Both total and methyl mercury have been found to increase in passive precipitation collectors following fires in northern Minnesota, with post-fire methyl mercury concentrations up to 8.75 times higher than the pre-burn average and even greater deposition in conifer-dominated areas (Witt et al. 2009).

These documented and predicted post-fire mercury increases raise concern over potential impacts to lakes, streams and wetland areas surrounding wildfires (Witt et al. 2009). Mercury concentrations in fish have been shown to increase following fires in Alberta, Canada (Kelly et al. 2006), by increasing mercury inputs and more importantly by restructuring food webs through greater productivity and increasing piscivory, with a 5-fold increase in whole-body mercury accumulation in rainbow trout (Oncorhynchus mykiss). Collectively, these studies suggest that

fire-related mercury emissions, and climate-related mercury mobilization from previously frozen ground, have the potential to impact aquatic biota, including piscivorous fish such as northern pike. Our data set will serve as a baseline for biomonitoring of mercury in Alaska's National Wildlife Refuges.

In summary, we created a substantive data set on which subsistence management and consumption decisions can be based for Yukon Delta, Innoko, Selawik, Koyukuk-Nowitna, and Yukon Flats NWRs. We met our ancillary goals, which included testing for correlation between mercury and methyl-mercury and establishing an average methyl-mercury:total mercury ratio in northern pike muscle; comparing mercury and methyl-mercury concentrations among regions, and comparing methyl-mercury concentrations to consumption thresholds. We have communicated the results to the communities which participated in the study, presented the results to statewide audiences, and worked with the Alaska Dept. of Health and Human Service – Section of Epidemiology to educate and inform subsistence users on NWRs about mercury in subsistence fish. A manuscript for publication in a peer-reviewed scientific journal that combines our data on mercury in pike with Alaska Department of Public Health data on hair mercury in human subsistence consumers from pike sampling villages is undergoing internal review by the State of Alaska.

Raw data are available upon request from angela\_matz@fws.gov.

#### MANAGEMENT RECOMMENDATIONS

Biomonitoring of contaminants, especially as they relate to human health concerns, often appears irrelevant to fish and wildlife managers. The FWS is responsible, however, for continuing to provide subsistence opportunities on National Wildlife Refuges in Alaska, and as land managers are responsible for warning resource users of potential hazards on Refuges. This project allows NWR managers to meet those responsibilities for mercury in pike, a well-used subsistence fish on NWRs in northern and western Alaska.

By using the State of Alaska's consumption limits for mercury in fish, managers can specifically identify villages (when sampled) that would benefit from communication and outreach regarding mercury in a commonly used subsistence food. We developed and performed this outreach in collaboration with our human-health partners at the State of Alaska, and have delivered it through village visits, mailings, and now online through the State of Alaska website to our partner NWRs. Refuge Managers can use our data and the consumption recommendations to clarify potential hazards for subsistence users of their Refuge. In particular, subsistence users living in the Yukon Delta National Wildlife Refuge have a better understanding of the hazards of mercury in subsistence fish.

Refuge managers also have a baseline data set for biomonitoring of mercury in Alaska's NWRs. As mercury deposition increases with increasing global industrialization, and as mercury mobilization and transport into biota potentially increases with climate change, these data can

and should form the basis of periodic monitoring on our National Wildlife Refuges in Alaska through the National Wildlife Refuges Inventoring and Monitoring program for Alaska.

#### **ACKNOWLEDGEMENTS**

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## APPENDIX A

Number of northern pike (*Esox lucius*) collected for mercury analysis, by sample site, collected on National Wildlife Refuges in Alaska, 2005-2007.

National Wildlife Refuge	Watershed Region	Sample Site	Number of pike
Yukon Delta	Kuskokwim	Gweek River	15
		Johnson River	32
		Tuluksak	13
		Aniak	15
		Whitefish Lake	15
		unspecified	13
Yukon Delta	Lower Yukon	Emmonak	7
		Atchuelinguk	25
		Kuyukutak River	15
		Paimiut Slough	15
		Andreafsky R	12
		Holy Cross	12
Innoko	Mid-Yukon	American Creek	15
		Innoko	9
		Hather Creek	13
		Kaiyuh Flats	10
Koyokuk/Nowitna	Mid-Yukon	Huslia	17
		Sulukna	13
Selawik	Northwest Alaska	Hannah Davis	17
		Tuglumaagruk	13
		Kiana	17
		Noatak	17
		Buckland	15
		Emma Ramoth	13
		Noorvik	12
Yukon Flats	Upper Yukon	Burman Lake	5
		Chandalar River	13
		Lower Birch	8
		Upper Birch	4
		Black River	15
		Birch Creek	9
		Dall River	15
		Ray River	1